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(54) **Method and apparatus for manufacturing fibre-reinforced material.**

(57) A method for impregnating fibre bundles with molten or liquid resin in the course of manufacturing a fibre-reinforced material (which material comprises one or more fibre bundles, wherein each fibre is surrounded by matrix resin) by impregnating a continuous web (10; 10a) of one or more fibre bundles with molten or liquid resin (32,33) and solidifying said resin by cooling or chemical reaction. During the impregnation the molten or liquid resin (35) material is subjected to shear forces by bringing it between two closely spaced surfaces (10; 10a, 36; 37) which are in moving relationship to each other.

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Method and apparatus for manufacturing fibre-reinforced material

The present invention concerns a method and apparatus for manufacturing a fibre-reinforced material. Specifically the present invention concerns a method and apparatus for manufacturing fibre-reinforced material, which is formed by a reinforcing fibre bundle or bundles bonded with a matrix forming resin material in such a way that also the individual fibres are surrounded by resin material.

The main problem in the manufacture of products of this kind is the high viscosity of some matrix forming materials. Because the individual fibres in the fibre bundles are very tightly near each other, it is thereby difficult for the resin material to penetrate into the bundle to surround all individual fibres. However this would be necessary in view of the properties of the product, such as strength, stiffness, chemical resistance etc.

Many methods have been tried for the solution of the problem. In one solution fibre bundles are transferred through a so-called cross-head die, in which molten or liquid resin is penetrated into the fibre bundles moving through the die. In the so-called powder method a web of fibre bundles is transferred through a bed of thermoplastic resin particles, whereby the web of fibre bundles carries along resin particles, which at a later stage are melted. There are also solvent methods in which fibre bundles are impregnated with resins dissolved in liquid medium.

The cross-head die method has proved to be ineffective in the impregnation of fibre bundles with thermoplastic resin because all individual fibres will not be surrounded by resin material. Powder methods have proven to involve a multiple of processing stages which are difficult to synchronize, are expensive to purchase and operate while damaging the reinforcement. In solvent methods a drying process is required, which is expensive, causes environmental problems, is time-consuming and limits the production rate. Furthermore the quality of the product is low due to voids left after the removal of solvents.

The present invention provides a method and apparatus for impregnating reinforcing fibres in the form of fibre bundles so that individual fibres are surrounded by bonding resin. The invention is based on the known phenomena that given pseudoplastic materials being made subject to shear stresses experience a decrease in viscosity. This phenomena, known as shear thinning, is common for various emulsions, dispersions, suspensions and other materials including melts of thermoplastic resins and solutions of thermoset resins. The tendency for shear thinning response can be altered with the level of shear stress applied to the material. At low stresses a little response is achieved, at intermediate shear stresses shear thinning the response increases dramatically, while at very high stresses the response reverts to lower levels as the material approaches minimum viscosity.

A typical range of viscosity for thermoplastic resin melts is 10^2-10^5 PaS and for thermoset resin solutions 100-5000 PaS. In the impregnation of reinforcing fibres a range of 1-10 PaS is ideal, a common magnitude is 10^2 PaS and in some cases 10^3 PaS can be acceptable. Typical shear rate ranges for polymer melts and solutions are 10^{-1}-10^5 1/S and the range of maximum response can be 10^0-10^4 1/S.

The method according to the invention is concerned with impregnating fibre bundles with molten or liquid resin in manufacturing a fibre-reinforced material, which material comprises one or more fibre bundles, wherein each fibre is surrounded by matrix resin, by impregnating a continuous web of one or more fibre bundles with molten or liquid resin and solidifying said resin by cooling or chemical reaction. The method of the invention is characterized in that during said impregnation said molten or liquid resin material is made subject to shear forces by bringing it between closely nearby surfaces which are in moving relationship into each other.

According to an advantageous embodiment of the method according to the invention one of said surfaces is a stationary surface and the other surface is a moving surface. However, both of said surfaces can be moving surfaces as well. The essential feature in view of the invention is that said surfaces are nearby each other and that they are in moving relationship to each other.

According to an advantageous embodiment of the method according to the invention one of the moving surfaces comprises the continuous web of a fibre bundle itself or bundles themselves to be impregnated. In this case the resin to be impregnated is lead between one fibre bundle to be impregnated and the stationary or moving surface, whereat the surfaces naturally are moving in different directions to each other. This causes shear forces that are directed to the resin to be impregnated and decrease the viscosity of the resin and facilitate the covering of the individual fibres with resin.

According to an advantageous embodiment of the method according to the invention the moving surfaces are cylindrical surfaces, in which case one of the surfaces is formed by the web to be impregnated. Said surfaces can also be planar surfaces.

wherein one of the surfaces can be the web to be impregnated itself or said surfaces can be formed of two discs that rotate in opposite directions and are nearby each other.

With the method of the invention it is possible to manufacture any fibre reinforced material, in which the fibres are in form of bundles. The fibres can be any kind of fibres bondable by a matrix resin. Such products are for example fibre-reinforced granules for injection molding or other processes, prepregs or preimpregnated materials for additional processes such as pultrusion, filament winding, tape laying etc.

Suitable fibres for the invention are thus glass fibres, carbon fibres and aramid fibres and thermoplastic fibres. Most common fibre materials to be used are glass fibre products in the form of bundles, which can be used as single bundles or also in the form of woven products, for example as glass fibre rovings. Typically glass fibre bundles contain thousands of individual fibres with a diameter of 10-17 µm.

The resin material to be used four bonding fibre-reinforced products can be a thermoplastic resin, which is impregnated into fibre bundles as melt, or a thermosetting resin, which will be solidified after impregnation with heat or chemical reaction.

Suitable thermoplastic resins are among others olefine homo and copolymers, vinyl chloride homo and copolymers, polyethylene terephtalate, acrylonitrile homo polymers and copolymers, polyamides or copolyamides, thermoplastic polymers of formaldehyde, polycarbonate, polysulphone and mixtures of two or more of any of the polymers above or in general any other thermoplastics which show decreased viscosity by shearing action.

The invention concerns also an apparatus for impregnating reinforcing fibres in the form of fibre bundles in manufacturing fibre-reinforced material, which material comprises one or more bundles, wherein each fibre is surrounded by matrix resin. The apparatus of the invention is characterized in that it comprises means for subjecting shear forces during said impregnation into said molten or liquid resin, said means consisting of two closely nearby surfaces, between which surfaces said molten or liquid resin material is forced to flow and thus made subject to shear forces during impregnation, and means for feeding said molten or liquid resin material between said surfaces.

According to an advantageous embodiment of the apparatus according to the invention at least one of said surfaces is a moving surface.

According to an advantageous embodiment of the apparatus according to the invention both of said surfaces are moving surfaces.

According to an advantageous embodiment of the apparatus according to the invention one of said surfaces comprises a web of a fibre bundle to be impregnated.

According to another embodiment of the apparatus according to the invention said surfaces are sylindrical or planar surfaces.

According to an advantageous embodiment of the apparatus according to the invention said means for feeding molten or liquid resin between said surfaces comprise one or more feed openings in the wall of an impregnation head, which openings have circular, rectangular or other cross-section.

According to an advantageous embodiment of the apparatus according to the invention said feed openings are one or more slits cut into the wall of an impregnation head.

According to an advantageous embodiment of the apparatus according to the invention said impregnation head is connected to the outlet of an extruder.

According to an advantageous embodiment of the apparatus according to the invention the impregnation head is curved or planar within the area od impregnation.

The invention will be further illustrated but not limited by referring to the enclosed figures, in which

Fig. 1 is a principal drawing of an apparatus according to the invention applied for manufacturing continuously fibre-reinforced thermoplastic granules.

Fig. 2 is an end view of an impregnation head according to the invention, where the surfaces being in moving relationship with each other are formed by the fibre bundle web itself and the outer surface of a cylindrical impregnation head.

Fig. 3 is an end view of an impregnation head according to the invention, where the surfaces being in moving relationship with each other are formed by the fibre bundle web itself and the outer surface of a rotating impregnation head.

Fig. 4 is an end view of an impregnation head according to the invention, where the surfaces being in moving relationship with each other are formed by the fibre bale web itself and the outer surface of the impregnation head, which outer surface is planar within the impregnation area.

Fig. 5 is a detail of an impregnation head showing several feed openings having a round cross-section for supplying molten thermoplastic resin to the impregnation area.

Fig. 6 is a detail of an impregnation head showing one slit-like feed opening for supplying molten thermoplastic resin to the impregnation area, and

Fig. 7 is a detail of an impregnation head

showing two slit-like feed openings having a round cross-section and opening to opposite sides of the impregnation head for supplying molten thermoplastic resin to the impregnation area.

In Fig. 1 a web 10 of glass fibre bundles is supplied from a drum 11 through a preheating chamber 12 and a spreading device 13 into an impregnation head 18. In the spreading device 13 the web 10 is transported via set of rolls 14, so that the fibre web runs in an alternating way over and under the rolls. This spreads the individual fibres in the bundles and thus facilitates the impregnation in the impregnation stage. However this spreading device 13 is not necessary and it does not form a part of the invention. The impregnation head 18 is connected to an extruder 15, into which the resin material 16 to be impregnated is fed through a hopper 17. In the extruder 15 the resin is melted and is formed to be ready to be fed to the impregnation head 18 according to the invention.

The resin impregnated web 10b or band of fibre bundles is then transferred to a cooling unit 19, where the molten resin is solidified by cooling. The solidifying can also take place by chemical reaction, whereat the impregnated resin may contain additives necessary for the solidifying and, in the solidifying, extra heating can naturally be used, if necessary.

From the cooling unit 19 the resin impregnated and solidified fibre bundles are then transferred to a chopper 20, which cuts the impregnated web to pieces 21 of proper length, which are ready for packing or are ready to be used for manufacture of the products. With the method and apparatus of the invention it is naturally also possible to prepare continuous fibre-reinforced products, which in that case are cut in pieces of wished lengths or are left completely uncut.

The transportation of the web 10 through the apparatus in Fig. 1 can be carried put by any suitable pulling device. A belt-driven pulling device 23 is schematically illustrated in Fig. 1.

In Fig. 2 there is disclosed an impregnation head 30, inside which there is a feed channel 31. The resin 32 to be impregnated is pumped through this channel 31 for example from an extruder (not shown), where the thermoplastic resin to be impregnated is melted. From the channel 31 a feed opening 33 extends to the cylinder-like outer surface of the impregnation head 30 at point 34. The web of fibre bundles 10a travels to the direction marked with arrow A past this impregnation point 34, whereby the resin is pulled between the surfaces of the web 10a and the surface 36 of the impregnation head 30 and thus made subject to shear forces, which lower its viscosity and ensures a maximum impregnation of the fibre bundles. The fibre bundle web impregnated with resin is marked with 10b.

Fig. 3 is similar like Fig. 2 except that the impregnation head 30 is formed by a rotating cylinder, which rotates to the direction of arrow B. In this embodiment the shearing action is stronger and takes place within a longer area as in the apparatus according to Fig. 2.

In the embodiment according to Figs. 2 and 3 the two surfaces, between which the shearing action takes place, have a cylinder-like form. In Fig. 4 the impregnation takes place between the planar surfaces 37 of the impregnation head 30 and the surface of web 10.

In Figs. 5 and 6 different shapes of the feed openings 33 has been disclosed. In Fig. 5 there are three openings 33 with a circular cross-section. There can be one ore several openings depending on the width of the fibre bundle web 10. In Fig. 6 there is one slit-like feed opening 33, the width of which is approximately same as the one of the fibre bundle web. In Fig. 7 there is disclosed an embodiment, in which there is two slit-like feed openings 33 to the opposite sides of the impregnation head 30. Of course there can be more than 2 slit-like feed openings. It is possible also in the embodiment of Fig. 5 to use two or more groups of feed openings 33.

Although the invention has been described above applied to the impregnation of amoving fibre bundle web, it is evident that the fibre bundle web can be stationary and the impregnation head can be arranged to be movable. The function of the invention will be same in both cases. Further it is possible to use two or more impregnation heads according to the invention and these impregnation heads can be placed on the same or different sides of the fibre web to be impregnated.

Furthermore it is possible to pretreat the resin to be impregnated in another such way in the method and apparatus of the invention, by which its viscosity is decreased.

Claims

1. A method for impregnating fibre bundles with a molten or liquid resin in the course of manufacturing a fibre-reinforced material (which material comprises one or more fibre bundles, wherein each fibre is surrounded by matrix resin) by impregnating a continuous web (10; 10a) of one or more fibre bundles with molten or liquid resin (32,33) and solidifying said resin by cooling or chemical reaction, characterized in that during said impregnation the molten or liquid resin (35) material is subjected to shear forces by bringing it between two closely spaced surfaces (10; 10a, 36; 37) which are in moving relationship to each other.

2. A method according to claim 1, characterized in that one of said surfaces is a stationary surface (36; 37) and the other surface is a moving surface (10; 10a).

3. A method according to claim 1, characterized in that both of said surfaces (10; 10a, 36; 37) are moving surfaces.

4. A method according to any of the preceding claims, characterized in that the moving surfaces or one of the moving surfaces comprises the continuous web (10; 10c) of a fibre bundle itself.

5. A method according to any of the preceding claims, characterized in that said surfaces are cylindrical surfaces (36) or planar surfaces (37).

6. A method according to any of the preceding claims, characterized in that said resin matrix is thermoplastic material, preferably comprising one or more of olefine homo and copolymers, vinyl chloride homo and copolymers, polyethylene terephthalate, acrylonitrile polymers and copolymers, polyamides or copolyamides, thermoplastic polymers of formaldehyde, polycarbonates or polysulphones.

7. An apparatus for the impregnation of fibre bundles with molten or liquid resin in the course of manufacturing fibre-reinforced material (which material comprises one or more fibre bundles, wherein each fibre is surrounded by matrix resin) characterized in that it comprises means for subjecting shear forces in the molten or liquid resin (35) during said impregnation, said means consisting of two closely spaced surfaces (10; 10a, 36; 37) being in moving relationship to each other, between which surfaces said molten or liquid resin material is caused to flow and thus subjected to shear forces during impregnation; and means (31,33) for feeding said molten or liquid resin (35) between said surfaces (10; 10a, 36; 37).

8. Apparatus according to claim 7, characterized in that at least one of said surfaces is a moving surface (10; 10a).

9. Apparatus according to claim 7, characterized in that both of said surfaces are moving surfaces (10; 10a).

10. Apparatus according to claim 7, characterized in that both of said surfaces are moving surfaces (10; 10a).

11. Apparatus according to any one of claims 7-10, characterized in that one of said surfaces comprises a web (10; 10a) of a fibre bundle to be impregnated.

12. Apparatus according to any one of claims 7-11, characterized in that said surfaces are cylindrical surfaces or planar surfaces (37).

13. Apparatus according to any one of claims 7-12, characterized in that said means for feeding molten or liquid resin between said surfaces comprise one or more feed openings (33) in the wall of an impregnation head (30), which openings (33) have a circular, rectangular or other cross-section.

14. Apparatus according to claim 11, characterized in that said feed openings (33) are slits cut in the wall of an impregnation head.

15. Apparatus according to claims 13 or 14, characterized in that said impregnation head (30) is connected to the outlet of an extruder (15).

16. Apparatus according to any one of claims 13-15, characterized in that the impregnation head is curved or planar within the area of impregnation.

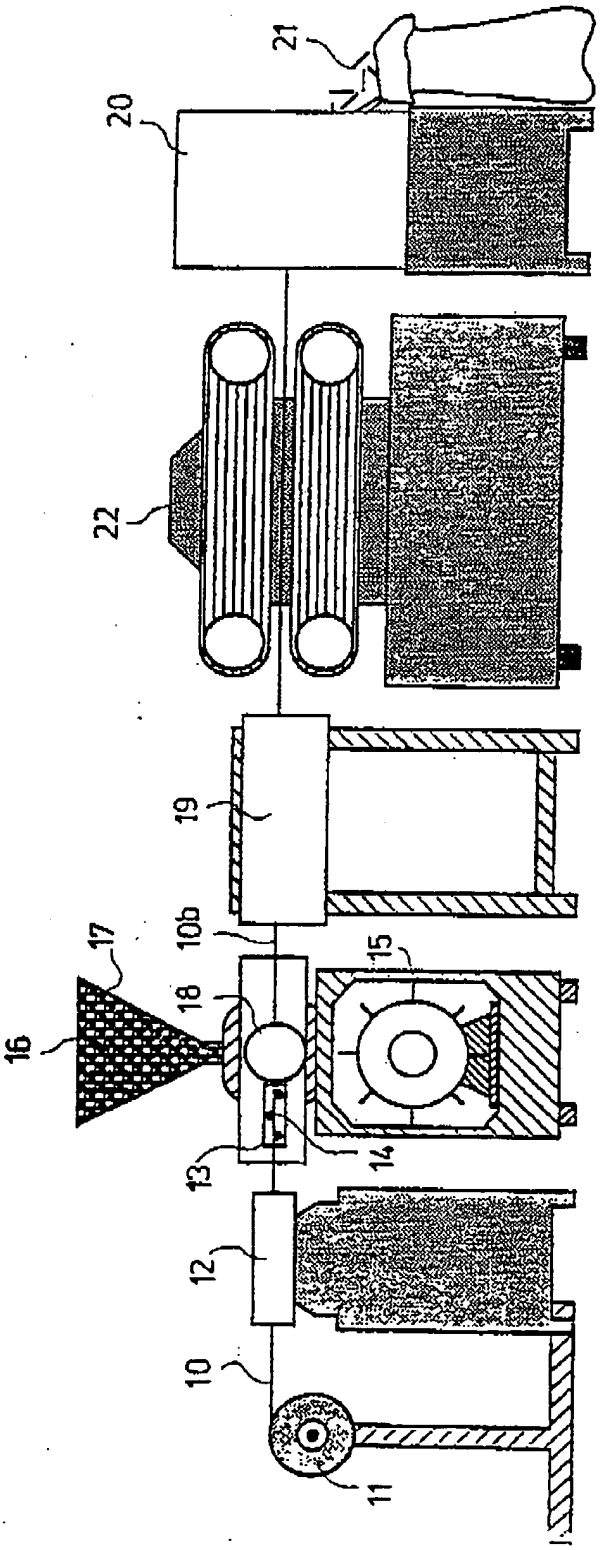

FIG. 1

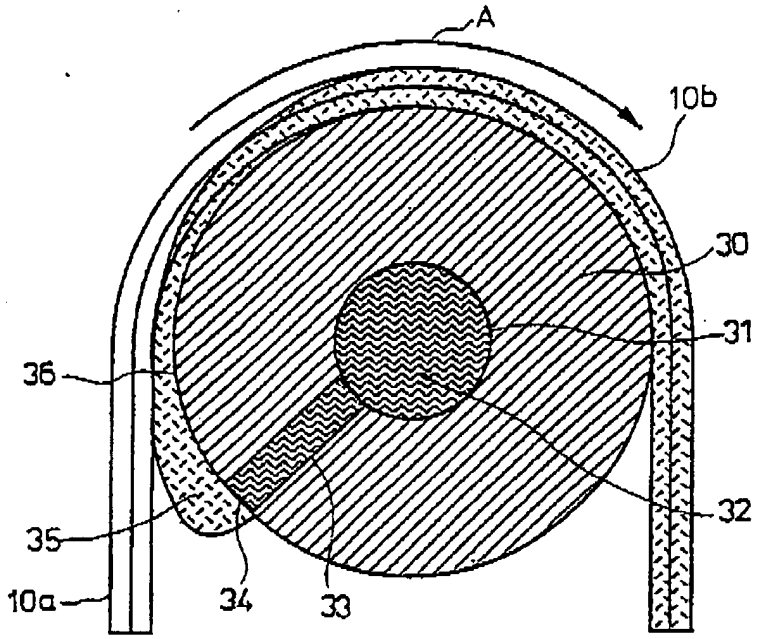

FIG. 2

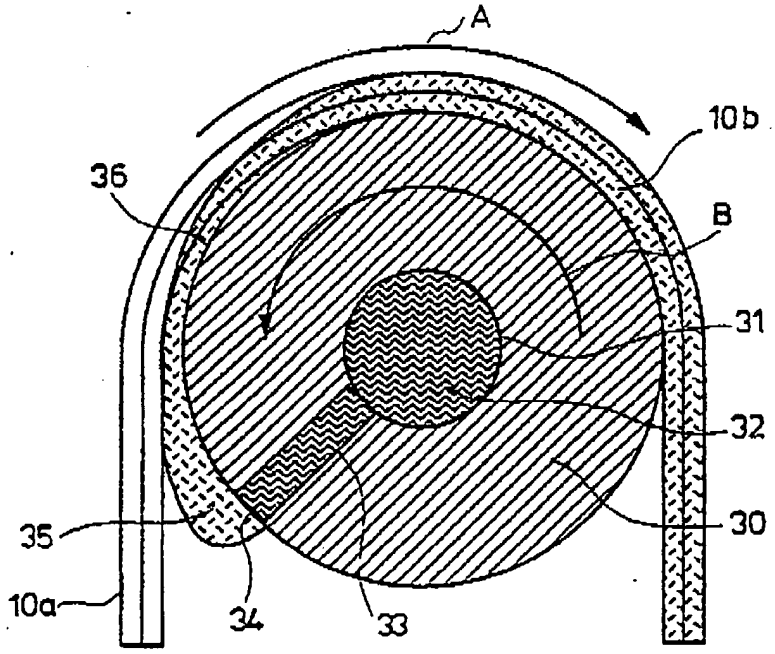

FIG. 3

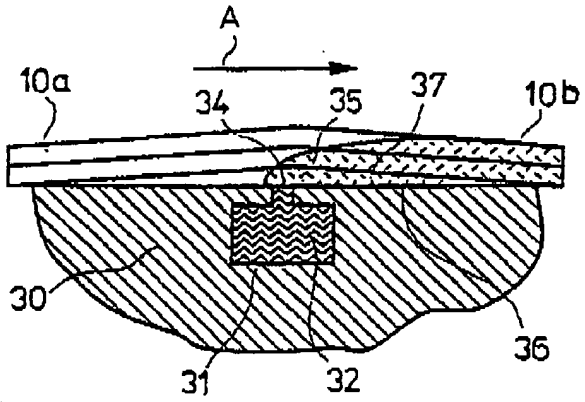

FIG. 4

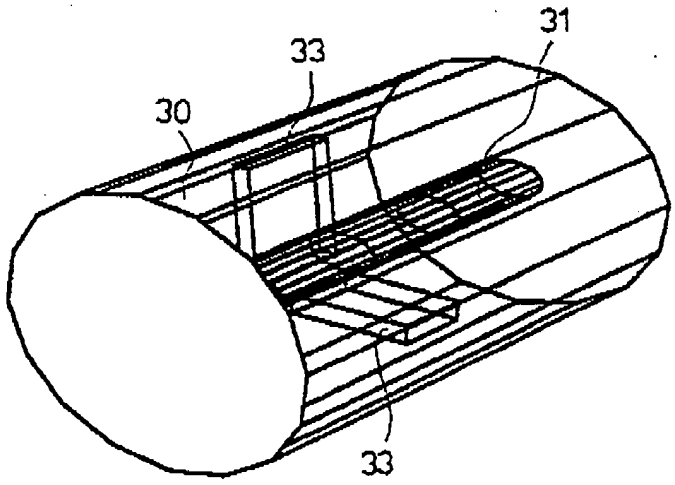

FIG. 7

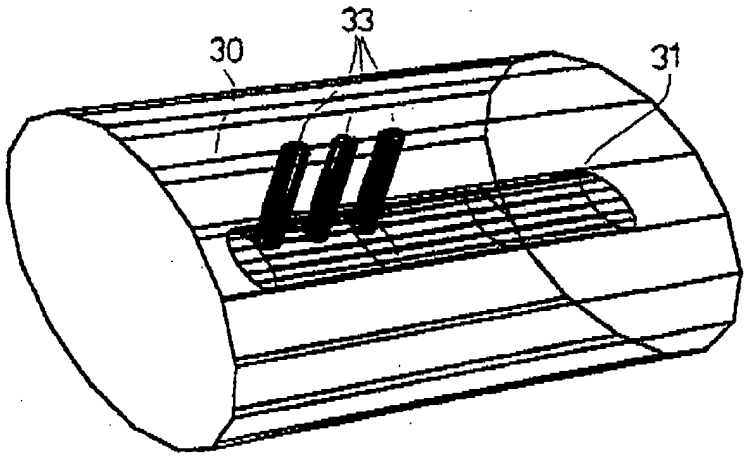

FIG. 5

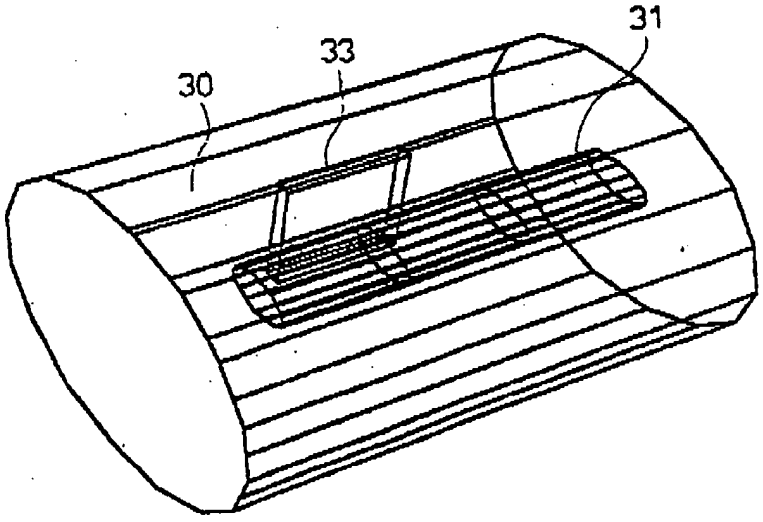

FIG. 6